

1 FORMABLE, POROUS, CHEMILUMINESCENT REACTANT COMPOSITION AND
2 DEVICE THEREFOR

3
4 FIELD OF THE INVENTION

5 This invention is directed toward the field of
6 chemiluminescent compositions and more particularly, to
7 devices which produce light from an immobilized
8 chemiluminescent material.

9
10 BACKGROUND OF THE INVENTION

11 The term "chemiluminescent reactant",
12 "chemiluminescently reactive" or "chemiluminescent reactant
13 composition" is interpreted to mean a mixture or component
14 thereof which will result in chemiluminescent light
15 production when reacted with other necessary reactants in
16 the processes as disclosed herein.

17 The term "fluorescent compound" is interpreted to mean
18 a compound which fluoresces in a chemiluminescent reaction,
19 or a compound which fluoresces in a chemiluminescent
20 reaction.

1 The term "chemiluminescent composition" is interpreted
2 to mean a mixture which will result in chemiluminescence.

3 The term "deagglomerate" is interpreted to mean to break
4 up or loosen a compacted portion of a cluster or a mass.

5 The term "fluidizable solid admixture" is interpreted to
6 mean a non-liquid admixture which behaves as a pseudo fluid
7 when agitated, but has properties of a solid when at rest.

8 Chemiluminescent light production generally utilizes a
9 two-component system to chemically generate light.
10 Chemiluminescent light is produced by combining the two
11 components, which are usually in the form of chemical
12 solutions referred to as the "oxalate" component and the
13 "activator" component. All suitable oxalate and activator
14 compositions, inclusive of the various additional
15 fluorescers, catalysts and the like, known to be useful in
16 the prior art, are contemplated for use within the present
17 invention.

18 The two components are kept physically separated prior
19 to activation by a variety of means. Often, a sealed,
20 frangible, glass vial containing one component is housed

1 within an outer flexible container containing the other
2 component. This outer container is sealed to contain both
3 the second component and the filled, frangible vial. Forces
4 created by intimate contact with the internal vial, e.g. by
5 flexing, cause the vial to rupture, thereby releasing the
6 first component, allowing the first and second components to
7 mix and produce light. Since the objective of this type of
8 device is to produce usable light output, the outer vessel
9 is usually composed of a clear or translucent material, such
10 as polyethylene or polypropylene, which permits the light
11 produced by the chemiluminescent system to be transmitted
12 through the vessel walls. These devices may be designed so
13 as to transmit a variety of colors by either the addition of
14 a dye or fluorescent compound to one or both of the
15 chemiluminescent reactant compositions or to the vessel.
16 Furthermore, the device may be modified so as to only
17 transmit light from particularly chosen portions thereof.

18 Examples of such a chemiluminescent system include:
19 U.S. Pat. No. 5,043,851 issued to Kaplan. Kaplan discloses
20 a polygonal, chemiluminescent lighting device which
21 concentrates light in the corners of the device, thus

1 enhancing visibility of light emanating from the light stick
2 portion of the device and optimizing the amount and
3 distribution of light radiated.

4 U.S. Patent No. 4,626,383 to Richter et al. discloses
5 chemiluminescent catalysts in a method for producing light
6 in short duration, high intensity systems, and low
7 temperature systems. This invention relates to catalysts
8 for two component chemiluminescent systems wherein one
9 component is a hydrogen peroxide component and the other
10 component is an oxalate ester-fluorescer component. Lithium
11 carboxylic acid salt catalysts, such as lithium salicylate,
12 which lower the activation energy of the reaction and also
13 reduce the temperature dependence of the light emission
14 process are taught.

15 U.S. Patent No. 5,121,302 to Bay et al. describes a
16 solid, thin, chemiluminescent device emitting light in one
17 direction. The device is comprised of a back sheet of a
18 laminated metal foil having heat sealed thereto at its edges
19 a bi-component front sheet and a temporary separation means
20 positioned to divide the interior area into two
21 compartments. The bi-component includes a first component

1 of which is a laminated metal foil and a second component of
2 which is a transparent or translucent polyolefin sheet. The
3 metal foil of the bi-component offers heat stability,
4 increased shelf life, and relative impermeability to
5 volatile components of the activator solution. The metal
6 foil laminate for activator solution storage enables the
7 activator solution to retain its viability due to the
8 impermeability of the metal foil.

9 U.S. Patent No. 6,062,380 to Dorney discloses a glow
10 cup system with illumination capabilities. The apparatus is
11 a generally cylindrically-shaped container made out of a
12 semi-rigid material, with a preferred embodiment comprised
13 of a translucent plastic material, to allow limited
14 flexibility at the outer layer of the cup as its form can be
15 somewhat altered temporarily by applying pressure to the
16 sides. Within the side wall of the cup is a cavity. The
17 cavity contains a plurality of rupturable ampoules
18 containing a chemiluminescent fluid. The chemiluminescent
19 fluid within the ampoule is an oxalate. A second
20 chemiluminescent fluid resides within the cavity so that
21 when the ampoule breaks open, the two fluids make contact

1 and provide illumination. The ampoule is broken by applying
2 pressure by the user on the outer layer of the cup at the
3 cavity point. The bottom of the cup contains a plug, which
4 may or may not be removable, which seals the second
5 chemiluminescent component within the cavity spacing.

6 Additionally, it is desirable to produce
7 chemiluminescent light from objects of various shapes or
8 forms. U.S. Pat. No. 4,814,949 issued to Elliott discloses
9 a means of making shaped, two-dimensional, chemiluminescent
10 objects. Conventional liquid, chemiluminescent reagents are
11 combined to produce light. A non-woven, absorbent article
12 in the desired shape is permitted to absorb the
13 chemiluminescent reagents after mixing and activation so
14 that the article emits light from the shape desired.
15 Although the shape may be as simple or as complex as
16 desired, it is essentially limited to a two-dimensional
17 surface and is additionally limited to producing a single
18 color of light per device.

19 An example of creating a chemiluminescent system
20 capable of producing light from a swellable polymeric
21 composition is disclosed in U.S. Pat. No. 3,816,325 issued

1 to Rauhut et al. Two primary means are employed to produce
2 solid chemiluminescent systems. The first system relies on
3 diffusion of a chemiluminescent oxalate solution into a
4 solid polymer substrate such as a length of flexible vinyl
5 tubing. The diffusion process occurs when a length of the
6 vinyl tubing is immersed in a suitable chemiluminescent
7 reagent for an extended period of time. After removal of
8 the tubing from the oxalate solution, application of liquid
9 activator to the surface of the tubing causes the tubing to
10 emit light. Since the solid polymer is relatively non-
11 porous, it is difficult to rapidly and completely activate
12 the oxalate in the tubing because the relatively slow
13 process of diffusion must also be relied upon to permit the
14 activator solution to reach the chemiluminescent reagent
15 diffused into the polymer before light can be generated.

16 In a further embodiment of U.S. Patent No. 3,816,325,
17 the chemiluminescent oxalate solution is mixed with a
18 polyvinyl chloride (PVC) resin powder to form a paste, which
19 is then spread on a substrate and baked in an oven to form a
20 flexible, elastic film. While this embodiment is operative,
21 the polyvinyl chloride sheet described exhibits weaknesses

1 in uniformity, strength, flexibility, and most importantly,
2 porosity. Additionally, the processes described are
3 primarily suitable for producing relatively thin objects
4 only.

5 U.S. Pat. No. 5,173,218 to Cohen et al. discloses a
6 combination of PVC polymer resins to produce a porous,
7 flexible, chemiluminescent structure from liquid slurries.
8 Although an improvement in the art, the products produced
9 still suffer from a variety of shortcomings, particularly if
10 solid, chemiluminescent objects are to be produced which are
11 other than relatively flat, thin objects. A thin "pad" is
12 produced from a mixture of polymer resins, which is strong
13 and flexible, and exhibits satisfactory absorptive
14 properties of the activator fluid. However, the processes
15 taught focus on producing pads which are made by pouring a
16 liquid slurry mixture into molds. As such, the slurry and
17 hence, the resulting pad shape, is limited to the shape of
18 the mold, into which the slurry is poured and pools.
19 Additionally, it is well-known to those skilled in the art
20 that the formulas and processes utilized in the prior art
21 may produce chemiluminescent pads with a relatively tough

1 and impermeable "skin" wherever the slurry has been in
2 contact with the mold during the baking process. This skin
3 is easily recognized as a darker and more transparent region
4 of the pad and is highly impermeable. Consequently, it is
5 incapable of rapidly absorbing liquid activator solution and
6 as such, minimally contributes to light output of the
7 device. The thickness of this skin varies with the time and
8 temperature of the baking process, but in any event, this
9 skin represents wasted material from which little usable
10 light may be produced. It has been determined that this
11 skin is created by an inability of the slurry to draw in air
12 (or other gasses) during the baking process. To achieve a
13 significantly porous product, air must enter the slurry
14 mixture during the baking process from the exposed surfaces
15 of the slurry pool. During the curing process, air is
16 usually drawn into the pad to replace the volume occupied by
17 solvents which become absorbed into the PVC resins. This
18 process continues as air is drawn down to ever increasing
19 depths within the pad as first the upper regions of the pad
20 cure and then successively lower regions of the pad cure.
21 It is this inclusion of air into the pad during the baking

1 process which primarily determines the percent of open pore
2 space and hence absorptiveness of the pad. At some point
3 during the baking process described, the bottom of the mold
4 may reach a temperature at which the slurry mixture in
5 contact with this region of the mold begins to jell and
6 cure, even though an air path from the exposed surfaces of
7 the slurry to this lower region may not have been created.
8 Due to a lack of air available to this jelling slurry, this
9 "bottom up" curing process results in a pad which is tough,
10 dense, and virtually non-porous in the region of the pad
11 proximal to the mold bottom and to a lesser extent, the mold
12 edges. Certain adverse effects of this bottom up curing
13 process can be minimized if the bottom of the mold is placed
14 on a cold thermal mass in the curing oven, thereby providing
15 for heating and curing of the bottom portion of the slurry
16 following the remainder of the slurry. Nonetheless, the
17 undesirable production of a tough and impermeable skin layer
18 remains unaddressed.

19 During the baking processes, such as those disclosed in
20 U.S. Patent No. 5,173,218, the slurry expands as air is
21 drawn into the polymer matrix, which air adds to the volume

1 of the matrix. As a result, significant problems develop
2 when attempting to cure a relatively large mass of this
3 slurry. For example, if a liquid slurry mixture, as taught
4 in the '218 patent, is poured into a test tube and baked for
5 the appropriate time to cure, a dense, tough mass will be
6 produced exhibiting very poor porosity and hence, poor
7 absorbency throughout most of the mass. This is due in part
8 to the "bottom up" curing process described above wherein
9 insufficient air is drawn into the slurry during the curing
10 process due to the existence of an air tight liquid layer
11 above the slurry being cured near the mold bottom.
12 Additionally, it has been unexpectedly found that the slurry
13 materials will not draw in the requisite air if the slurry
14 is inhibited from expanding during the curing process. In
15 the case of the test tube example above, the side walls of
16 the test tube constrain the slurry from expanding and
17 drawing in the air required to produce a cured matrix with
18 the high degree of porosity and absorbency required to
19 permit activation of the product with liquid activator.
20 Even though the slurry is free to expand vertically in the
21 test tube during the curing process, the lateral constraint

1 on the slurry by the walls of the test tube is sufficient to
2 prevent optimal expansion of the slurry and air induction
3 into the mass during the curing process. As such, the cured
4 mass will exhibit low porosity and yield poor light output
5 which is a limitation of the art.

6 It is often desirable to provide a chemiluminescent
7 device which is not only capable of producing light, but
8 producing light in a variety of colors. U.S. Patent No.
9 5,508,893 issued to Nowak et al. is directed toward a multi-
10 color chemiluminescent lighting device and method of
11 producing the product. This device is comprised of a
12 flexible tube filled at least partially with an activator
13 solution, a plurality of ampoules containing oxalate
14 solutions located within the tube, and at least one barrier
15 element between ampoules to prevent color mixing. This
16 device is capable of imparting different chemiluminescent
17 colors following activation.

18 U.S. Patent No. 5,705,103 issued to Chopdekar et al.
19 describes a composition for producing chemiluminescent light
20 of controllable duration. The composition is comprised of
21 an oxalate component (including an oxalate ester) in a

1 solvent, an activator component (a peroxide compound and a
2 catalyst) in a solvent, and a fluorescer. By appropriate
3 selection of the molecular weight of the homopolymer for the
4 oxalate component, control of the total glow time and the
5 point in time at which commencement of light production
6 occurs may be varied. Although this device provides a
7 controllable duration or stability of light, there is no
8 suggestion of a composition to control the generation of gas
9 produced or a composition which may be independent of a
10 container, i.e. not formable or porous.

11 Thus, what is lacking in the art is a means for
12 producing three-dimensional objects which are self-
13 illuminated by means of chemiluminescence, and producing a
14 highly porous composition to exhibit quick activation and
15 excellent light output. In addition, the prior art fails to
16 contemplate a product which may be independent of a
17 container, minimizes dark areas due to gas generation, and
18 which is capable of generating a plurality of spatially
19 separated or wavelengths of chemiluminescent light
20 simultaneously.

21

1 SUMMARY OF THE INVENTION

2 The instant invention teaches a means to produce three-
3 dimensional objects which are self-illuminated. The objects
4 may be as simple or as complex as desired. The objects are
5 produced by a method employing a formable, chemiluminescent
6 reactant composition. This composition is of such a nature
7 that it may be readily placed in variously shaped containers
8 and then cured in said containers whereupon the composition
9 becomes solid and is of a shape that precisely matches the
10 container in which it was formed. Once formed, the
11 composition is semi-rigid and may be removed from the
12 container if desired. Additionally, the instant invention
13 provides for a chemiluminescent reactant composition which
14 is exceptionally porous and is not limited to relatively
15 flat strips of material, as is the case in prior art. Also,
16 objects produced by means of the instant invention may be
17 hollow so that a minimal amount of material may be used to
18 produce glowing, three-dimensional objects. Further, these
19 objects may be multi-colored, that is, a single object can
20 be created which is capable of simultaneously generating a

1 plurality of spatially separated colors or wavelengths of
2 chemiluminescent light.

3 A fundamental objective of the instant invention is
4 that a significant portion of the interstitial spaces in the
5 solid product necessary for quick and reliable activation by
6 a liquid activator is created prior to curing. As such, the
7 system does not rely primarily on porosity created during
8 the curing process in which the air must enter the matrix
9 from outside. Since the final porosity of the product of
10 the instant invention is primarily a function of the degree
11 of densification prior to curing, the final porosity of the
12 product may be precisely and advantageously controlled.

13 Typically, it is desirable to provide a product which
14 will activate as quickly as possible. For this to occur,
15 the activator solution must react quickly and completely
16 with the oxalate portion of the chemiluminescent system.
17 There are times, however, when it may be desirable to slow
18 down the rate of reaction or perhaps at least, slow down the
19 rate at which the activator is able to reach the oxalate
20 component and react with it. Since the product of the
21 instant invention may be densified to practically any

1 desirable degree, the interstitial space available through
2 which the activator communicates with the solid product may
3 be reduced as desired, thereby reducing the mobility of the
4 activator and its ability to react with the solid oxalate
5 containing component. Additionally, because most of the
6 porosity of the chemiluminescent solid is determined by the
7 degree of densification prior to curing, the product of the
8 instant invention can be cured in a relatively confined
9 space, such as a test tube, and the resulting product will
10 be highly porous and receptive to activator solution.

11 Accordingly, it is an objective of the instant
12 invention to provide for a means to produce three-
13 dimensional objects which are capable of self-illumination
14 through chemiluminescence and which objects may generate a
15 plurality of spatially separated colors or wavelengths
16 simultaneously.

17 It is a further objective of the instant invention to
18 produce a three-dimensional chemiluminescent object which is
19 highly porous.

20

1

2 It is a further objective of the instant invention to
3 provide for a three-dimensional chemiluminescent object in
4 which the porosity can be readily and precisely controlled.

5 It is a still further objective of the invention to
6 provide for a three-dimensional chemiluminescent object
7 which may be produced by forming in a mold which may not
8 permit significant expansion of the chemiluminescent
9 reactant composition during the curing process.

10 It is yet another objective of the instant invention to
11 provide for three-dimensional chemiluminescent objects which
12 have little or no dark regions due to "skin" effect caused
13 by improper curing.

14 It is a further objective of the instant invention to
15 provide for a three-dimensional chemiluminescent object
16 which may be formed in such a manner that the object is
17 hollow.

18 It is a still further objective of the invention to
19 provide for a three-dimensional chemiluminescent object in
20 which a substantial portion of the porosity is created prior
21 to the curing process.

1 It is yet another objective of the instant invention to
2 provide for a formulation for a chemiluminescent reactant
3 composition which is formable and as such, may be readily
4 formed into a desired shape either with or without the use
5 of a mold or form.

6 Other objectives and advantages of this invention will
7 become apparent from the following description taken in
8 conjunction with the accompanying drawings wherein are set
9 forth, by way of illustration and example, certain
10 embodiments of this invention. The drawings constitute a
11 part of this specification and include exemplary embodiments
12 of the present invention and illustrate various objectives
13 and features thereof.

14
15 BRIEF DESCRIPTION OF THE FIGURES

16 FIG. 1 is a chart depicting light output versus activation
17 time for differing bulk densities of solid oxalate;
18 FIG. 2 is a pictorial view of an example embodiment of the
19 instant invention;

1 FIG. 3 is a cross sectional view of the example embodiment
2 of Figure 2 illustrating placement of the chemiluminescent
3 reactant composition;
4 FIG. 4 is a cross sectional view of Figure 3 illustrating
5 densification of the chemiluminescent reactant composition
6 using a tamping tool;
7 FIG. 5 is a cross sectional view of the example embodiment
8 following densification, illustrating second
9 chemiluminescent reactant component ampoule placement and a
10 void in the fluidizable solid admixture;
11 FIG. 6 is a cross sectional view of another example
12 embodiment of the instant invention illustrating placement
13 of the chemiluminescent reactant composition;
14 FIG. 7 is a cross sectional view of the embodiment of Figure
15 6 illustrating positioning of a compression tool within the
16 chemiluminescent reactant composition;
17 FIG. 8 is a cross sectional view of an embodiment of the
18 instant invention illustrating densification of the
19 chemiluminescent reactant composition by the compression
20 tool of Figure 7; and,

1 FIG. 9 is a cross sectional view of an embodiment of the
2 instant invention illustrating the densified
3 chemiluminescent reactant composition.

4

5 DETAILED DESCRIPTION OF THE INVENTION

6 The present invention is directed toward a formulation,
7 process of making, and device for use regarding a
8 chemiluminescent reactant composition which is formable, and
9 may be used to produce a multi-dimensional object. This
10 composition overcomes weaknesses of the prior art and
11 implements the use of a novel forming process to be applied
12 to a chemiluminescent material, thereby furnishing a highly
13 porous, uniquely shaped, chemiluminescent object. The
14 process of the instant invention is not limited to the
15 conventional casting process producing relatively thin, flat
16 objects described in the prior art.

17 The formable and porous powder of the instant invention
18 may be readily compacted to various degrees, and upon heat
19 curing, may form a relatively strong, flexible, and highly
20 porous mass. The apparent density of a material is easily
21 controlled through the degree of compaction, or

1 densification. Therefore, an object of any desirable
2 apparent density may be produced. Because apparent density
3 directly affects the speed of activator absorption, the rate
4 of chemiluminescent activation may be advantageously
5 controlled.

6 Now with reference to the figures, Figure 1 is a chart
7 depicting how the control of bulk product density may be
8 used to vary activation time. Two devices were produced and
9 tested, each comprising a chemiluminescent reactant
10 composition which is in the form of a solid oxalate
11 containing composition, hereinafter referred to as a solid
12 oxalate. The first device had a bulk density of
13 approximately of 0.54 g/cc and reached maximum light output
14 at approximately 10 minutes after activation. The second
15 device, with a bulk density of about 0.72 g/cc, reached its
16 peak light output at approximately 37 minutes after
17 activation. This data indicated that activation time is
18 affected by bulk density with more compacted objects
19 requiring a longer period to activate. The ability to
20 control the light output curve enables the production of

1 chemiluminescent devices to meet a wide array of market
2 needs.

3 By way of example, for the production of large
4 chemiluminescent objects, a hollow chemiluminescent shape
5 may be preferred to a solid one, since there is a
6 diminishing return effect as light produced from deep within
7 the solid shape reaches the surface inefficiently and may
8 not be emitted as useful light. Additionally, a hollow
9 chemiluminescent shape provides for a convenient and elegant
10 means to introduce a second reactant component to the
11 product. An ampoule or vessel containing a second reactant
12 component may be placed inside the void in a hollow shape.
13 When the ampoule or vessel is ruptured, the second component
14 is readily absorbed by the interior surface of the hollow
15 shape and is quickly transferred by capillary action through
16 the porous, chemiluminescent matrix until the entire mass is
17 wetted and producing light through chemiluminescence.
18 Placing the second component means inside the void also
19 hides it from view and permits production of a more
20 aesthetically pleasing product.

21

1 An example of a form which may be produced using the
2 teaching of the instant invention is that of a
3 chemiluminescent candle. Such candles provide a safe,
4 reliable alternative to real candles. The flame from real
5 candles can ignite other objects. Unlike conventional
6 candles, chemiluminescent candles are wind resistant and
7 waterproof and by employing the instant invention, can be
8 produced to emit light in any color desired or in any
9 combination of colors or wavelengths from a single device.

10 Previous attempts at producing these "candles" which
11 use chemiluminescent systems as light sources have met with
12 drawbacks. Typically, a chemiluminescent lighting device,
13 such as a light stick, which employs liquids, has a head-
14 space in the device which represents approximately 30% of
15 the container volume. Light cannot be produced in this
16 head-space area. Japanese Pat. Application No. 10-170263
17 discloses an air bubble capture means in which the gaseous
18 head-space (or bubble) which is above the liquid
19 chemiluminescent fluid in a sealed chemiluminescent device
20 is trapped in a region of the device other than the
21 uppermost portion. By displacing the bubble from the upper

1 tip portion of a sealed chemiluminescent device, such as a
2 candle for example, the entire portion of the candle flame
3 tip will appear to glow during the chemiluminescent
4 reaction. If the bubble had been permitted to remain at the
5 flame tip, it would create a dark region near the top of the
6 flame since the area of the bubble will not produce any
7 light. Such a dark region would detract from the overall
8 visual acceptability of the device. Carbon dioxide, carbon
9 monoxide, and oxygen are common gases liberated in
10 peroxy luminescent systems. These gases rise to the top of
11 any liquid chemiluminescent system and form bubbles at the
12 top of the device. Thus, while the device described in
13 Japanese Pat. Application No. 10-170263 may effectively
14 eliminate the problem of bubbles initially contained at the
15 top of a chemiluminescent device, a method is not provided
16 to displace bubbles which are generated during the
17 chemiluminescent process. The instant invention permits a
18 candle or any other chemiluminescent object desired to be
19 produced in which an initial head-space bubble in the device
20 and any significant visible build up of bubbles in the
21 device as the chemiluminescent process progresses is

1 eliminated. In addition, the instant invention does not
2 require any specially formed traps, channels, or valves in
3 the device to realize this benefit. Since the formable mass
4 of the chemiluminescent system of the instant invention is a
5 solid, there is no space for bubbles to aggregate and
6 combine. While gasses generated during the chemiluminescent
7 process are still produced, these gasses are constrained
8 from rising in the solid formable mass and are evenly
9 distributed throughout the solid, which subsequently result
10 in a seemingly flawless output of light.

11 Figure 2 sets forth a preferred embodiment of the
12 present invention as a chemiluminescent candle 10 which
13 comprises a blow-molded envelope in the shape of a
14 candlestick. When the device is activated, the flame
15 portion of the candle glows.

16 A candle envelope 11, as illustrated in Figure 3, may
17 be created by blow-molding or other suitable forming means
18 from materials such as, but not limited to, polyethylene or
19 polypropylene. Preferably, the distal end of the candle
20 envelope opposite the flame shape is left open. The candle
21 envelope 11 is positioned so that the open end is up. A

1 formable chemiluminescent reactant composition 12 of the
2 instant invention, is placed into a candle envelope 11 so
3 that the envelope is partially full. While the fluidizable
4 solid admixture is flowable, it also exhibits a degree of
5 cohesiveness, and yields a packable, formable, moist powder.
6 Therefore, an auxiliary feeding means, such as a vibratory
7 feeder, may be useful to aid in feeding the formable
8 chemiluminescent reactant composition 12. Once the formable
9 chemiluminescent reactant composition 12 is in the candle
10 envelope 11, it may be compacted slightly with a tamping
11 tool 13, as illustrated in Figure 4, designed for this
12 purpose. This compression process not only serves to assist
13 the composition in conforming to the shape of candle
14 envelope 11, but also densifies the composition and
15 compresses it so that it will not flow or be further
16 displaced in the candle envelope 11 should the envelope
17 orientation be altered. The composition may however, be
18 removed from the envelope, if desired, by application of
19 sufficient vibratory forces so as to cause liquefaction of
20 the compacted chemiluminescent reactant composition 15. A
21 tamping tool 13 may be designed with a tapered tip 14 such

1 that it will not only compact the composition but also
2 produce a cavity 16, as illustrated in Figure 5, in the
3 resultant compacted composition. The cavity 16 provides a
4 convenient means to facilitate distribution of a second
5 chemiluminescent reactant component 18, such as within an
6 ampoule 17 and promotes quick, even activation of the
7 device. Once the second chemiluminescent reactant component
8 is in place, a plug 19 at the distal end of the candle
9 envelope 11 may be heat sealed. Additionally, the cavity 16
10 provides a space into which the composition may expand
11 during the curing process so that an exceptionally porous
12 product may be produced. The cavity is not required to
13 produce products which are highly porous but may be employed
14 in certain cases to produce products with exceptional
15 porosity. Such cavities are not possible with the processes
16 taught in the prior art.

17 As set forth in Figure 6, a chemiluminescent rose-
18 shaped envelope 21 is produced by first blow-molding an
19 envelope, from polyethylene by way of example, into the
20 shape of a rose bud with a stem attached. The diameter of
21 the stem is considerably smaller than that of the bud. For

1 this preferred embodiment, it is desirable to produce a rose
2 bud where the entire surface of the bud is illuminated
3 through chemiluminescence. It is also desirable to produce
4 the item using the least chemiluminescent material possible
5 which will create the desired effect. The rose-shaped
6 envelope 21 is filled with a small quantity of formable
7 chemiluminescent reactant composition 12.

8 Referring now to Figure 7, inserted into the rose-
9 shaped envelope 21 with a chemiluminescent reactant
10 composition 12 is a compression tool 22 comprising a hollow
11 needle 23 equipped with an expandable bladder 24, which for
12 purposes of illustration is depicted as being held in place
13 by at least one retainer ring 25. The distal end of the
14 hollow needle 23 is plugged and a hole in the side of the
15 needle beneath the expandable bladder 24 permits air
16 pressure from within the needle to fill and inflate the
17 expandable bladder 24. The bladder expands, as illustrated
18 in Figure 8, by using air pressure for inflation purposes,
19 whereby the formable chemiluminescent reactant composition
20 12 surrounding the inflated bladder 24 compacts against the
21 interior wall of the rose bud envelope. Figure 9

1 illustrates the compacted chemiluminescent reactant
2 composition 15 in a semi-solid state. Following this
3 compression process, the bladder deflates and the needle
4 probe is removed, leaving a cavity 16. The compacted
5 chemiluminescent reactant composition may subsequently be
6 cured by baking while in place inside the rose bud envelope,
7 in a preferred embodiment, at 95°C for 10 minutes. After the
8 composition cools, a sealed ampoule containing a solution of
9 second chemiluminescent reactant component is inserted into
10 the rose-shaped envelope 21 and a plug may be fitted to the
11 stem and heat sealed to form a hermetic seal as previously
12 described for the candle embodiment. The resulting product
13 is an object appearing as a realistic rose bud which, when
14 activated, emits light from the entire surface of the bud.
15 Activation is accomplished, by way of example, by simply
16 flexing the stem of the rose to fracture the ampoule and
17 release the second component which is then absorbed into the
18 chemiluminescent reactant composition, or formable solid
19 admixture. Since the compacted chemiluminescent reactant
20 composition highly conforms with the interior wall of the

1 envelope, even fine details such as petals of the rose are
2 captured by the process of the instant invention.

3 Concerning the candle and rose embodiments as
4 discussed, it is assumed that the cured solid product will
5 remain in the polymer envelope, however, the material may
6 just as easily be cast and cured in a mold and then removed.
7 Solid chemiluminescent objects may be produced employing the
8 instant invention using, by way of example, compression or
9 centrifugal molding. Individually shaped items produced by
10 the process of the instant invention could be included as
11 free-floating objects which would glow if placed in a vessel
12 containing a second chemiluminescent reactant component
13 solution. Such a system could produce, by way of example, a
14 "snow globe" which contains glowing snow particles. Since
15 the formable mass of the chemiluminescent reactant
16 composition in the instant invention is in solid form, a
17 plurality of positionable and spatially fixed colors may be
18 employed in a single device. For example, a rose bud may be
19 produced in which the bud is red with orange stripes.

20 A significant advantage of the instant invention over a
21 fully liquid chemiluminescent system, such as that found in

1 conventional light sticks, is that the entire surface of the
2 object may be caused to glow if desired.

3 Since the resultant product of the instant invention is
4 a solid chemiluminescent material, the product may be
5 utilized in situations where it is impractical or impossible
6 to use a liquid chemiluminescent system which is dependent
7 upon its container.

8 The following examples describe the experimental
9 process performed to reach the novelties of the present
10 invention.

11 A series of experiments were devised to identify
12 optimal materials and formulas necessary to produce a
13 formable, porous, chemiluminescent reactant composition. As
14 taught in the prior art, a pre-slurry may be prepared by
15 dissolving approximately 2 parts PVC resin (Geon Corp. #121)
16 with 98 parts of a chemiluminescent reactant solution which
17 is exemplified herein as an oxalate solution. A slurry was
18 prepared, also according to '218, by mixing 59 parts oxalate
19 pre-slurry (from above) with 31 parts medium particle size
20 PVC powder resin (Geon #218) and 9 parts large particle size

1 PVC resin (Geon #30). The resultant material is a pourable,
2 liquid slurry.

3 Examples 1-6

4 Six tests were conducted to determine the effects of
5 varying cure times and temperatures as well as slurry
6 thickness on porosity. In each test, approximately 7 grams
7 of liquid slurry was placed in a small aluminum weighing pan
8 which was then positioned on a spacer such that the pan
9 bottom was slightly inclined to create a slurry having a
10 depth ranging from 0.015" to 0.180". During each test, the
11 pan was placed on a wire rack in a circulating air oven.
12 After curing for the specified time, each sample item was
13 removed from the pan, sectioned, and examined for proper
14 curing and porosity. A properly cured sample is defined as
15 one in which all of the oxalate solution had been absorbed
16 into the PVC matrix, and which does not exhibit signs of
17 over-curing. In a properly cured matrix, the lower
18 molecular weight PVC particles fuse together. However, the
19 higher molecular weight PVC particles, while absorbing the
20 liquid oxalate solution, do not significantly fuse together.
21 If the curing time and temperature is excessive, the higher

1 molecular weight PVC particles will fuse together, resulting
2 in a matrix which is over-cured as evidenced by the presence
3 of dark and/or shiny regions within the cured sample,
4 referred to as a pad. This over-cured matrix will exhibit
5 very low porosity.

6 Table I illustrates the results obtained using various
7 slurry curing conditions:

8 Table I - Slurry Curing

Test Item	Cure Time (minutes)	Cure Temp (°C)	Result/Observation
1	3	95	Under-cured/doughy in thick areas
2	5	95	Possibly over-cured; non-porous in area which contacted pan
3	8	95	Non-porous in pan contact areas to about 1/3 thickness of pad
4	10	95	Dense; non-porous in bottom 1/3 thickness of pad
5	20	95	Dense; non-porous in bottom 1/3 thickness of pad
6	8	82	Less dense areas; dark and dense in areas which contacted pan

9
10 In test item 1, it was apparent that the PVC particles
11 had not completely absorbed the oxalate solution as the
12 material was doughy and contained significant amounts of
13 free liquid. In tests 2-5, the material was found to be
14 less doughy, but only the exposed surface of each cured
15 sample was determined to be porous.

1 Each of the test items 1-5 were activated with
2 chemiluminescent activator reagent. Items 2-5 glowed from
3 the surface, but did not produce significant light from the
4 dark, non-porous areas. Item 1 produced very little light
5 over most of its surface, presumably because the liquid
6 oxalate solution that was not absorbed into the PVC matrix
7 during the curing process presented a barrier, preventing
8 the activator solution from reaching the balance of the
9 liquid oxalate below the surface. Some glow was evident
10 near the surface of the matrix at the boundary layer where
11 the activator and oxalate solutions combined. Item 6 was
12 cured at a lower temperature since items 1-5 appeared to be
13 over-cured by application of excessive heat, causing the
14 high molecular weight PVC particles to fuse together. The
15 test results from item 6, however, contradicted this theory.
16 Even at the lower time and temperature used to cure item 6,
17 there was still evidence of dark, dense regions where the
18 pad had been in contact with the pan.

19

20

21

1 Example 7

2 A chemiluminescent candle was produced using the same
3 liquid slurry formation as that used in tests 1-6 above. To
4 make this candle, approximately 3.2 grams of liquid slurry
5 was injected into a polyethylene candle envelope using a
6 syringe. A glass ampoule containing chemiluminescent
7 activator was inserted into this slurry such that the lower
8 end of the ampoule contacted the inside bottom of the candle
9 envelope. The assembly was placed in a circulating air oven
10 set for 82°C and allowed to cure for 12 minutes. After
11 removal, the assembly was allowed to cool to room
12 temperature whereupon the candle envelope and cured slurry
13 were sectioned for observation. The PVC matrix (cured
14 slurry) appeared to be fully cured, but was dark and dense.
15 The PVC matrix portions were removed from the envelope and
16 placed in an aluminum weighing pan. Chemiluminescent
17 activator reagent was added to these portions whereupon the
18 cured slurry glowed dimly. It was observed that the
19 activator was reacting only with the outermost surface of
20 the cured slurry and could not reach the cured slurry
21 interior. This lack of absorption of the activator solution

1 into the cured slurry was determined not to be the result of
2 over-curing or under-curing of the slurry, but was due to
3 very low porosity exhibited by the pad. Porosity, or pore
4 space, within the matrix derives from two sources. A small
5 portion of this porosity results from the already porous PVC
6 particles in the matrix. A more significant factor in
7 determining the resultant porosity of the cured slurry is
8 the ability of air to be inducted into the entire slurry
9 volume during the curing process. It was observed when
10 utilizing the liquid slurry formulation taught in '218, that
11 if heat sufficient to cure the slurry reaches an interior
12 region before outer regions are fully cured and porous, the
13 interior region will cure with low porosity. This effect is
14 due to the inability of air to migrate through this
15 surrounding liquid region to the interior.

16 Example 8

17 With these results in mind, an aliquot of slurry was
18 supported upon an air permeable substrate, e.g. a 10 cm by
19 10 cm section of 2 mm thick non-woven polyester felt and
20 placed in a circulating air oven, maintained at 82°C, for 8
21 minutes. The expectation was that the felt would provide

1 uniform access of air to the slurry and that heat would cure
2 the slurry from the outside in such a manner that no dark,
3 non-porous regions would be formed as had been the case in
4 slurry previously cured in the impermeable aluminum pans.
5 As each successive layer of the slurry cured from the
6 outside in, it would become porous, thereby allowing air to
7 reach subsequent layers. This sample was removed from the
8 oven and allowed to cool. Upon inspection, it was noted
9 that the pad had no dark or dense areas and was extremely
10 porous. The sample was activated with chemiluminescent
11 activator reagent whereupon the sample glowed brightly and
12 evenly throughout its entirety.

13 A model which explains the formation of interstitial
14 spaces in PVC particle/solvent slurries is that in which
15 large, roughly spherical, PVC particles are joined together
16 by smaller, lower molecular weight, PVC particles to form a
17 matrix. The PVC particles absorb the solvent that had
18 initially filled the interstitial spaces between these
19 particles. If air is permitted to enter the matrix during
20 this curing process, the PVC particles will swell and expand
21 as the solvent is absorbed into the particles.

1 Example 9

2 To determine if increased air access through the slurry
3 could be achieved by using a greater weight percent of
4 larger particle PVC, a new preparation of slurry was created
5 and tested. This new slurry contained 56 parts pre-slurry,
6 29 parts medium size particle resin (Geon #218) and 15 parts
7 large particle size resin (Geon #30). Approximately 2.5 ml
8 of this liquid slurry was placed in a polyethylene candle
9 envelope to which a glass activator ampoule was added. The
10 item was cured at 75°C for 12 minutes and allowed to cool.
11 It was apparent after dissection that the slurry had been
12 cured, but still contained dark regions that were nonporous.

13 The theory that a formulation of PVC resin and liquid
14 oxalate could be produced in a manner that would result in a
15 material allowing air to move through the formulation at all
16 times prior to and during curing was then developed and
17 tested.

18

19 Example 10

20 A new formulation was created which utilized the pre-
21 slurry described above by dissolving approximately 2 parts

1 PVC resin (Geon Corp. #121) with 98 parts of an oxalate
2 solution. Although in this example the liquid oxalate
3 solution was propylene glycol dibenzoate based, any base
4 compound in the art is contemplated. In this new
5 formulation, a higher weight percent of a single PVC
6 particle was used in place of the medium and large particle
7 PVC resins employed in the slurries previously described.
8 Approximately 40 parts of pre-slurry were added to 60 parts
9 of resin (Geon #466). The resulting composition was not a
10 liquid slurry, but rather a moist, packable and formable
11 powder characterized as a fluidizable solid admixture. The
12 resin should be selected so as to contain a particle size or
13 range thereof sufficient to provide said fluidizable solid
14 admixture. In an illustrative, albeit not limiting
15 embodiment, this resin is a PVC resin having an average
16 particle size distribution of about 125 microns.

17 A large variety of polymers may be employed in the
18 polymeric composition: polyethylene, polypropylene,
19 poly(vinyl Chloride), Poly(methyl methacrylate), poly(vinyl
20 benzoate), poly(vinyl acetate), cellulose poly(vinyl
21 pyrrolidone), polyacrylamide, epoxies, silicones, poly(vinyl

1 butyral), polyurethane, nylons, poly acetyl, polycarbonate,
2 polyesters and polyethers are non-limiting examples. Cross-
3 linked polymers may also be employed, such as polystyrene-
4 poly(divinyl benzene), polyacrylamide-
5 poly(methylenebisacrylamide), polybutadiene-copolymers, and
6 the like. For most applications the polymer should be
7 selected in conjunction with the activating hydrogen
8 peroxide liquid so as to be dissolvable, swellable, or
9 otherwise permeable to said activating liquid. Such
10 permeability is normally desired to permit efficient contact
11 between the activating liquid, the chemiluminescent
12 material, and (when desired or necessary) the fluorescer.
13 It will often be desirable to select the polymer and
14 activating liquid so as to provide a particular diffusion
15 rate and thus control the intensity and duration of light
16 emission. Some useful polymer-solvent combinations are: 1)
17 poly(vinyl pyrrolidone)-water, 2) poly(vinyl styrene-
18 polydivinyl benzene) copolymer-ethylbenzene, 3) poly (vinyl
19 chloride-ethyl benzoate), 4) poly(methyl methacrylate
20 dimethyl phthalate). The permeability of polymers to
21 solvents is, of course, well known to the art and it is a

1 straightforward matter to select useful polymer/solvent
2 combinations. Solvents used as plasticizers are
3 particularly advantageous. It is not necessary for either
4 the chemiluminescent material or a fluorescer to be
5 soluble in the polymer itself, although where the polymer
6 does not itself provide solubility for both these
7 ingredients, the activating liquid should provide at least
8 partial solubility. Alternatively the polymer could be
9 plasticized with a solubilizing plasticizer.

10 The moist powder of the resulting chemiluminescent
11 reactant composition has a consistency similar to light
12 brown sugar. Due to the cohesive nature of the fluidizable
13 solid admixture, it has been found to be beneficial to
14 deagglomerate or loosen any compressed portions by a method
15 such as by being sifted through a screen mesh or stirred
16 with a whisking tool to insure that the moist powder would
17 not become compacted prior to use. To aid in material
18 placement, a vibratory feed system may be used as well.
19 Although the above serve as examples for loosening compacted
20 portions, any means for deagglomerating the fluidizable
21 solid admixture may be used. The newly created formulation

1 along with the discovery that pre-existing interstitial
2 spaces in the material are critical to the curing process
3 resulted in an immediate and significant improvement in the
4 time required for complete chemiluminescent activator
5 reagent absorption and corresponding light output.

6 The formable, chemiluminescent reactant composition
7 thus comprises a first chemiluminescently reactive component
8 in combination with an amount of first polymeric resin
9 particles effective to yield a uniform dispersion,
10 visualized as a liquid slurry. An amount of second
11 polymeric resin particles in combination with the uniform
12 dispersion in an amount effective to yield a fluidizable
13 solid admixture is then provided. This fluidizable solid
14 admixture may be molded to form a specific shape. A means
15 for deagglomerating the fluidizable solid admixture may be
16 provided in order to loosen any portions of the mass which
17 may have been compacted during preparation. A means to cure
18 the fluidizable solid admixture may also be provided either
19 with or without the use of a mold. In a preferred
20 embodiment, the first polymeric resin particles and second
21 polymeric resin particles are each a polyvinyl chloride

1 resin. Although an activator solution is commonly added to
2 a composition to commence the emission of light, the oxalate
3 and activator of the instant invention may be
4 interchangeable. In such a case the first
5 chemiluminescently reactive component might comprise an
6 oxalate, and the second chemiluminescently reactive
7 component might then comprise an activator. Optionally, the
8 first chemiluminescently reactive component might comprise
9 the activator and the second chemiluminescently reactive
10 component might then comprise an oxalate.

11 In order to provide a chemiluminescent system, the
12 second component must be included. Therefore, a
13 chemiluminescent composition of the present invention
14 comprises a first chemiluminescent reactant including a
15 first chemiluminescently reactive component in combination
16 with an amount of first polymeric resin particles effective
17 to yield a uniform dispersion and an amount of second
18 polymeric resin particles in combination with the uniform
19 dispersion in an amount effective to yield a fluidizable
20 solid admixture. A second chemiluminescent reactant
21 component is included, wherein contact between the first and

1 second chemiluminescent reactant components will result in
2 the generation of light. The generation of light includes
3 at least one distinct wavelength, within the visible or
4 invisible spectrum. A means may be provided to controllably
5 activate the fluidizable solid admixture.

6 A multi-dimensional chemiluminescent device is also
7 disclosed comprising at least one first chemiluminescent
8 reactant including a first chemiluminescently reactive
9 component in combination with an amount of first polymeric
10 resin particles effective to yield a uniform dispersion and
11 an amount of second polymeric resin particles in combination
12 with the uniform dispersion in an amount effective to yield
13 a fluidizable solid admixture. At least one fluidizable
14 solid admixture is dispersed within a multi-dimensional
15 container, whereby densification of the fluidizable solid
16 admixture causes the formation of the multi-dimensional
17 chemiluminescent device. Contacting the device with a
18 second chemiluminescent reactant component will result in
19 generation of chemiluminescent light. As previously noted,
20 the resulting emission of light may be of more than one
21 distinct wavelength or color. Means for compacting or

1 densification of the fluidizable solid admixture thereby
2 provide a means to controllably activate the fluidizable
3 solid admixture and may be accomplished by a variety of
4 techniques all contemplated by the instant invention. By
5 way of example, the densification of the fluidizable solid
6 admixture is by a molding technique, wherein a moldable
7 object is formed or a hollow object is formed having areas
8 of controlled densification. These variations in density
9 are illustrative of a controlling parameter for the light
10 emitting reaction, and result in an object which is termed
11 as being controllably activated.

12 A process for the production of a chemiluminescent
13 reactant composition of the instant invention comprises
14 providing a first polymeric resin, then combining a first
15 chemiluminescently reactive component, typically in solution
16 form, with an effective amount of the first polymeric resin
17 to create a slurry. A second polymeric resin is provided
18 which is combined with the slurry in an amount effective to
19 create a fluidizable solid admixture. A means for providing
20 controllable activation of the fluidizable solid admixture
21 is also included which may be accomplished by compacting the

1 admixture to a desired degree. As illustrated in Figure 1,
2 the more compact the mass, the longer it takes to reach a
3 peak light output.

4 This fluidizable solid admixture is significantly
5 different than the liquid slurry taught in U.S. Pat. No.
6 5,173,218 in that it is not a liquid and will not seek its
7 own level. The fluidizable solid admixture is also
8 significantly different from the paste described in U.S.
9 Pat. No. 3,816,325 in that it is flowable, but will neither
10 sag nor slump. Most significantly, this powder composition
11 has an intrinsically high degree of porosity and
12 interconnecting, interstitial air spaces. Additionally, the
13 fluidizable solid admixture has a cohesive nature which
14 permits it to be formed into definite, solid shapes by
15 simply pressing the moist powder together with a gentle
16 force. By way of example, the material may be manipulated
17 with a human hand, or placed between two plates to create a
18 thin sheet. Furthermore, the cohesion exhibited by the
19 moist powder is sufficient to retain a desired shape after
20 pressing. By way of example, the fluidizable solid
21 admixture may be pressed into small cakes either with or

1 without the use of forms and simply cured by baking in an
2 oven whereupon the individual particles in the powder bond
3 together into a single porous mass. In an alternative
4 embodiment, the fluidizable solid admixture may be placed
5 into a mold and baked (cured) to form a solid object which
6 has a shape precisely matching that of the mold. Since the
7 moist powder, once slightly compacted, is not flowable such
8 as a dry powder or liquid slurry would be, the fluidizable
9 solid admixture of the instant invention may be shaped,
10 processed, or otherwise manipulated in such a manner that a
11 hollow object is produced. Such hollow chemiluminescent
12 objects have great value in that the outer, light-emitting
13 surface of the object may be formed into any desired shape
14 while maintaining a hollow interior. This hollow interior
15 not only permits conservation of chemiluminescent material
16 and thereby reduces cost, but also allows relatively large
17 chemiluminescent objects to be produced which exhibit a high
18 surface brightness at minimal cost.

19 Although PVC is the preferred polymeric resin, the
20 polymeric composition is not limited thereto.

1 Various methods for shaping and/or processing are
2 applicable to the chemiluminescent reactant composition of
3 the present invention. Examples of such methods include,
4 but are not limited to, injection molding, extrusion,
5 compression molding, cast molding, powder molding, or
6 electrostatic deposition, such as xerography. Powder
7 molding comprises dry blending the moist powder and a
8 curable additive to form a moldable composition.

9 Additionally, the fluidizable solid admixture may be
10 deposited electrostatically through a process such as
11 xerography, wherein the surface of a container retaining the
12 chemiluminescent reactant composition is given an electric
13 charge. Adhesion between the chemiluminescent reactant
14 composition and the container surface occurs only at the
15 charged areas to enable particular placement of a
16 chemiluminescent reactant composition within a container.

17 All patents and publications are herein incorporated by
18 reference to the same extent as if each individual
19 publication was specifically and individually indicated to
20 be incorporated by reference.

1 It is to be understood that while a certain form of the
2 invention is illustrated, it is not to be limited to the
3 specific form or arrangement herein described and shown. It
4 will be apparent to those skilled in the art that various
5 changes may be made without departing from the scope of the
6 invention and the invention is not to be considered limited
7 to what is shown and described in the specification and
8 drawings/figures. One skilled in the art will readily
9 appreciate that the present invention is well adapted to
10 carry out the objectives and obtain the ends and advantages
11 mentioned, as well as those inherent therein. The
12 embodiments, methods, procedures and techniques described
13 herein are presently representative of the preferred
14 embodiments, are intended to be exemplary and are not
15 intended as limitations on the scope. Changes therein and
16 other uses will occur to those skilled in the art which are
17 encompassed within the spirit of the invention and are
18 defined by the scope of the appended claims. Although the
19 invention has been described in connection with specific
20 preferred embodiments, it should be understood that the
21 invention as claimed should not be unduly limited to such

1 specific embodiments. Indeed, various modifications of the
2 described modes for carrying out the invention which are
3 obvious to those skilled in the art are intended to be
4 within the scope of the following claims.

5

6

7

8

9

10

11

12

13

14

15

16

17

18

19

20

21